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Oscillation of the tunnel splitting induced by temperature in the biaxial nanospin system

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Employing an instanton approach, we study the temperature dependence of the tunnel splitting in the biaxial spin system with a magnetic field along the hard axis. We show that for a given magnetic field the tunnel splitting oscillates and becomes topologically quenched with increasing temperature. We find that the topological oscillation becomes more prevalent in nanospin systems with larger basal anisotropy. This feature is expected to be observable in nanomagnets including Fe_8 molecular magnets. [S0163-1829(99)50830-8]

A quest to understand the macroscopic quantum tunneling of single domain ferromagnetic particles has been a topical issue of intensive theoretical and experimental studies over the past few years.[1] Magnetic molecular clusters of Mn_{12} acetate and Fe_8 have been such good candidates because all the clusters are identical with no dispersion on the size of the clusters and the number of interacting spins, and the spin ground state and the magnetic anisotropy are known with great accuracy.[2,3] Even though strong experimental evidence exists for quantum tunneling of the magnetization in Mn_{12} acetate,[2] several basic issues including the existence of the nondiagonal interactions remain unclear. However, Fe_8 clusters possess the strong transverse anisotropy which induces the tunnel splitting Δ between the two degenerate levels of each molecule and shows a pure quantum regime below 360 mK.[3]

The tunnel splitting of the ground state for the biaxial spin system has been studied by several groups.[4] Among them, the topologically quenched tunnel splitting in the biaxial spin system with a magnetic field was studied by Garg.[5] Using the quantum phase interference suggested by Loss, DiVincenzo, and Grinstein,[6] and by von Delft and Henley,[7] he showed that Δ oscillates with increasing external magnetic field applied along the hard anisotropy axis, and the topological freezing or unfreezing of tunneling have nothing to do with Kramer degeneracy. Such oscillations have been reported by Wernsdorfer and Sessoli in Fe_8 clusters.[8] Very recently, Chudnovsky and Martínez Hidalgo[9] have found that the switching from oscillations to the monotonic growth of Δ exists beyond the field range studied in Ref. 5, due to the cancellation between the real-time motion of the instanton and the contribution of the topological phase. Also, the meaning of the previously discussed oscillation of the splitting has been clarified by Garg.[10] Theoretical studies have been focused on the tunnel splitting of the ground states in the presence of the magnetic field along the hard axis at zero temperature. In this paper we consider the tunnel splitting of the biaxial spin system in the presence of temperature and magnetic field. We show that for a given magnetic field along the hard axis, the tunnel splitting oscillates with increasing temperature and it is topologically quenched quasiperiodically. This feature can be tested in experiments on nanomagnets including Fe_8.

Consider the spin coherent state path-integral representation of the partition function given by

$$Z(\beta\hbar)=\oint D[\mathbf{M}(\tau)]\exp(-S_E/\hbar), \tag{1}$$

where $\beta=1/k_BT$, the path sum is over all periodic paths $\mathbf{M}(\tau)=\mathbf{M}(\tau+\beta\hbar)$, and S_E is the Euclidean action which includes the Euclidean version of the magnetic Lagrangian L_E as

$$S_E=\int_0^{\beta\hbar}\left\{ i\frac{M}{\gamma}[1-\cos\theta(\tau)]\frac{d\phi(\tau)}{d\tau}+E[\mathbf{M}(\tau)]\right\}d\tau \tag{2}$$

in the spherical coordinates of the magnetization $\mathbf{M}$. Introducing the biaxial anisotropy with a magnetic field along the hard axis whose form is $E=k_1M_z^2+k_2M_y^2-MH_z$, we have, up to an additive constant,

$$E=K_1(\cos\theta-\cos\theta_0)^2+K_2\sin^2\theta\sin^2\phi, \tag{3}$$

where $\cos\theta_0=H/H_c$, $H_c=2K_1/M$, $K_1(=k_1M^2)>K_2(=k_2M^2)>0$ are the anisotropy constants, and x, y, and z are taken as the easy, medium, and hard axis, respectively. The first term in Eq. (2) which is called the Wess-Zumino-Berry phase term has a geometrical meaning,[11] and is of crucial importance in the ensuing discussion.

Performing the Gaussian integration over $\cos\theta$ in Eq. (1), we have the effective action given by

$$S_E^{\rm eff}=i\hbar S\int_0^{\bar\tau_p}d\bar\tau\left(1-\frac{h}{1-k\sin^2\phi}\right)\frac{d\phi}{d\bar\tau} +\hbar S\sqrt{k}\int_0^{\bar\tau_p}d\bar\tau\left[\frac{1}{2}m(\phi)\left(\frac{d\phi}{d\bar\tau}\right)^2+U(\phi)\right], \tag{4}$$

where $S=M/\hbar\gamma$, $k=K_2/K_1$, $\bar\tau=\omega_0\tau$, $h=H/H_c$, $\omega_0=2\gamma\sqrt{K_1K_2}/M$, and

$$m(\phi)=\frac{1}{1-k\sin^2\phi}, \tag{5}$$

$$U(\phi)=\frac{1}{2}\sin^2\phi\left(1-\frac{h^2}{1-k\sin^2\phi}\right). \tag{6}$$

As is shown in Fig. 1, there are three different ranges of the field, in which the position $\phi=\pi/2$ is the maximum for $h<1-k$, becomes the local minimum for $1-k<h<\sqrt{1-k}$,

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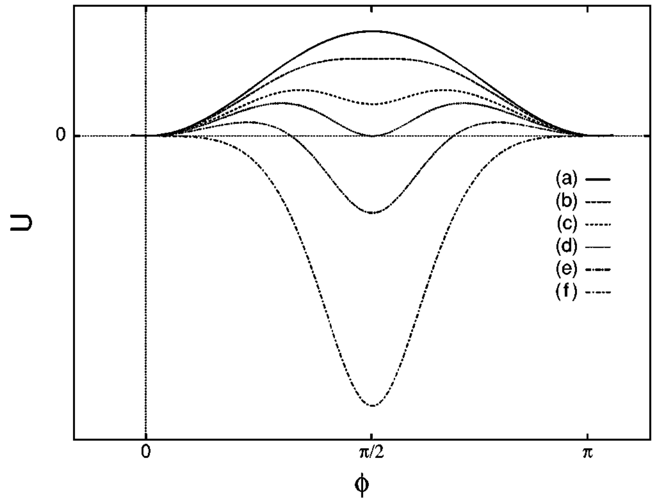

FIG. 1. The potential $U(\phi)$, where (a) $h<1-k$, (b) $h=1-k$, (c) $1-k<h<\sqrt{1-k}$, (d) $h=\sqrt{1-k}$, (e) $\sqrt{1-k}<h<1$, and (f) $h=1$.

and the global minimum for $\sqrt{1-k}<h<1$. The maximum starts to change from $\pi/2$ to either $\phi_m[=\arcsin\sqrt{(1-h)/k}]$ or $\pi-\phi_m$ at $h=1-k$ and vanishes at $h=1$ which defines the critical field, H_c. In this paper, we will focus on the behavior of the tunnel splitting in the field region $h<1-k$ at finite temperature.[12] In this situation the particle of mass $m(\phi)$ moves from ϕ_3 to $\phi_4(=\pi-\phi_3)$ or ϕ_2 to $\phi_1(=-\pi-\phi_2)$ in an inverted potential $-U(\phi)$, as is shown in Fig. 2. Using the energy conservation

$$\frac{1}{2}m(\phi)\left(\frac{d\phi}{d\bar{\tau}}\right)^2-U(\phi)=-E, \tag{7}$$

the first integral of Eq. (4) for these paths becomes

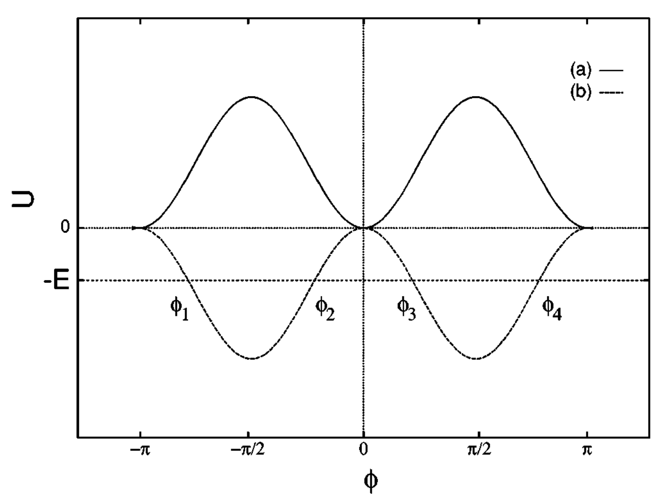

FIG. 2. The shape of the potential $U(\phi)$ in (a) and the inverted potential $-U(\phi)$ in (b), where the field range is $h<1-k$, and ϕ_i's ($i=1,2,3,4$) are the intersection points of $-U(\phi)$ and the particle energy $-E$.

$$\frac{1}{\hbar}S_E^{\text{eff, I}}=\pm S\pi\Bigg\{1-\frac{h}{\sqrt{1-k}}-\frac{2}{\pi}\times\left[\phi_3-\frac{h}{\sqrt{1-k}}\arctan(\sqrt{1-k}\tan\phi_3)\right]\Bigg\}, \tag{8}$$

where $\phi_3=\arcsin\sqrt{x_-}$ and x_- is defined in Eq. (10). Adding these two contributions, we have the factor $|\cos\Phi|$ for the tunnel splitting $D\exp(-S_E^{\text{eff, R}}/\hbar)|\cos\Phi|$, where D is related with the fluctuation determinant and $\Phi(H,E(T))=S_E^{\text{eff, I}}/\hbar$. Thus, the tunnel splitting is expected to be quenched at the condition satisfying $\Phi=(n+1/2)\pi$, which is determined by changing the temperature as well as the magnetic field. The second integral of Eq. (4) for either $\phi_3\to\phi_4$ or $\phi_2\to\phi_1$ becomes

$$\frac{1}{\hbar}S_E^{\text{eff, R}}=S\Bigg\{\frac{2x_-}{\sqrt{x_+-x_-}}[\Pi(\alpha^2,q)-\Pi(\alpha_3^2,q)+kx_+(\Pi(\alpha_3^2,q)-\mathcal{K}(q))]+\sqrt{k}\left(\frac{T_0}{T}\right)E\Bigg\}, \tag{9}$$

where $T_0=\hbar\omega_0/k_B$ and the parameters are

$$x_\pm=\frac{1-h^2}{2k}+E\pm\sqrt{\left(\frac{1-h^2}{2k}+E\right)^2-\frac{2E}{k}}. \tag{10}$$

Also, $\mathcal{K}$ and Π are complete elliptic integrals of the first and third kind with $\alpha^2=1-x_-$, $q^2=(1-x_-)x_+/(x_+-x_-)$, and $\alpha_3^2=(1-x_-)/(1-kx_-)$.[13]

Since the tunnel splitting depends on the temperature T via the energy E, we need to know the relation between E and T. In order to do that, we use the periodicity $\bar{\tau}_p$ of the particle in the range $\phi_3\to\phi_4$ ($\phi_2\to\phi_1$). From the energy conservation (7), we have

$$\frac{T}{T_0}=\frac{\sqrt{k(x_+-x_-)}}{2\mathcal{K}(q)}, \tag{11}$$

whose approximate form becomes

$$E\simeq\frac{8(1-h^2)^2}{1-h^2-k}\exp\left[-\frac{T_0}{T}\sqrt{1-h^2}\right], \tag{12}$$

for $q\lesssim 1$, i.e., in the limit of small T. Until now, the tunnel splitting Δ and the temperature T are the function of the energy E, and thereby the thermal behavior of Δ can be obtained by varying E in the energy range $0\leqslant E\leqslant U(\pi/2)[=(1-k-h^2)/(2(1-k))]$. At this point, we need to specify the crossover temperature T_c from the thermal to quantum regime because the first- or the second-order transition can occur around T_c. In the case of the first-order transition we should study nonmonotonic dependence of $\bar{\tau}_p$ on E to determine T_c. Meanwhile, if it is the second-order

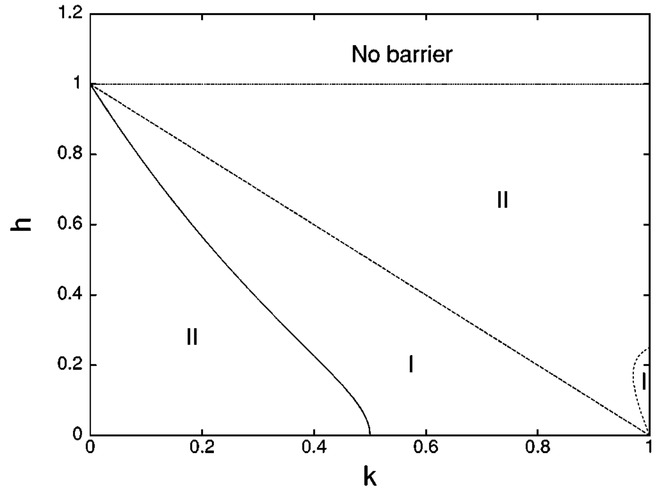

FIG. 3. The phase diagram h vs k, where I and II indicate the first- and the second-order transition, respectively. See the text for details.

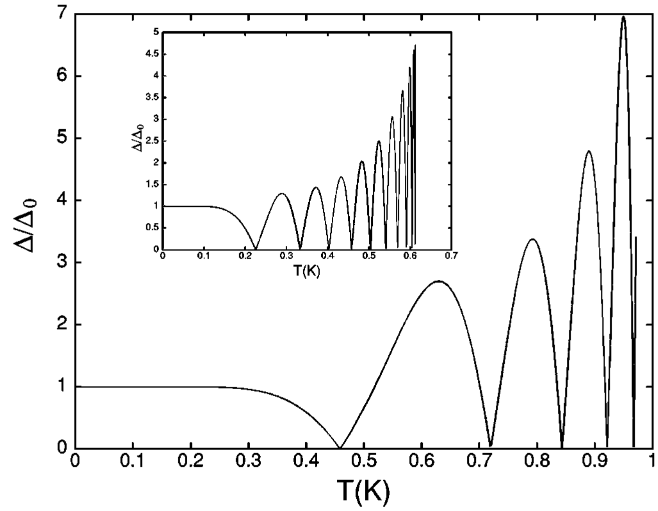

FIG. 4. The scaled tunnel splitting Δ/Δ_0 as a function T, with $S=10$, $k=0.713$, $h=0.02$, and $T_0=5.42$ K for Fe_8, where Δ_0 is the tunnel splitting at $T=0$ and the transition is first order. Inset: the same parameters except $k=0.1$ and $T_0=2.03$ K, where the transition is second order.

transition, $E=U(\pi/2)$ provides T_c in Eq. (11), which is represented as

$$\frac{T_c}{T_0}=\frac{1}{\pi}\sqrt{1-k-\frac{h^2}{1-k}}. \tag{13}$$

Here we note that the crossover temperature decreases with increasing field. In order to determine the first- or the second-order transition, we consider the behavior of $S_E^{\mathrm{eff,R}}$ around the top of the barrier. Expanding the integrand in Eq. (4) near ϕ_b which corresponds to the top of the barrier, and introducing the energy variable[15] p $[\equiv(U_{\max}-E)/\Delta U]$ and $\Delta U=U_{\max}-U_{\min}$, where $U_{\max}$ ($U_{\min}$) corresponds to the top (bottom) of the potential, the effective action (9) becomes

$$\frac{1}{\hbar}S_E^{\mathrm{eff,R}}(p)=\frac{\Delta U}{k_BT}[1+\alpha p+\beta p^2+O(p^3)], \tag{14}$$

where $\alpha=T/T_0^{(c)}-1$ with $T_0^{(c)}=\sqrt{|U''(\phi_b)|/m(\phi_b)}/(2\pi)$ and the criterion parameter which determines first- or second-order is given by[16]

$$\beta=\frac{\Delta U}{16U_2}\left\{\frac{12U_4U_2+15U_3^2}{2U_2^2}+3\left(\frac{m'(\phi_b)}{m(\phi_b)}\right)\left(\frac{U_3}{U_2}\right)+\frac{m''(\phi_b)}{m(\phi_b)}-\frac{1}{2}\left(\frac{m'(\phi_b)}{m(\phi_b)}\right)^2\right\}, \tag{15}$$

with the derivatives of the potential denoted by $U_2=-U''(\phi_b)/2(>0)$, $U_3=U^{(3)}(\phi_b)/3!$, and $U_4=U^{(4)}(\phi_b)/4!$. Using the analogy with the Landau model of phase transition, the factor α changes signs at the phase transition temperature $T=T_0^{(c)}$. If the factor β is negative (positive), the system becomes the first- (second-) order transition. Noting that the value of ϕ_b changes depending on whether the field range is $h<1-k$ or $h>1-k$, one sees from Fig. 3 that there exist two regimes which exhibit the first-order transition, and the phase boundaries are given by $h_{(<)}=(1-k)\sqrt{(1-2k)/(1+k)}$ and

$$h_{(>)}^{\pm}=\frac{16-16k+k^2\pm k\sqrt{k^2+32k-32}}{16-2k}, \tag{16}$$

where $<$ ($>$) indicates the field region h smaller (larger) than $1-k$. Thus, the first-order region is surrounded by $h_{(<)}<h<1-k$ and $h_{(>)}^{-}<h<h_{(>)}^{+}$.

We now calculate the tunnel splitting by numerically solving the results (8), (9), and (11) up to the preexponential factor. Taking the measured value of the anisotropy parameter $k=0.713$ for Fe_8, it is seen from Fig. 3 that the system exhibits the first-order transition in the range of field $h<1-k$. The oscillations are evident in Fig. 4. Even though it is quasiperiodic, it is remarkable that the tunnel splitting is quenched at several temperatures, which is originated from the topological phase term. Another interesting point is that the tunnel splitting initially decreases monotonically with increasing temperature and switches from monotonic to oscillatory behavior. This can be understood from the fact that, since the factor $|\cos\Phi|$ oscillates as a function of T via $\phi_3[T(E)]$, a small change of E near $E=0$ induces a large change of T close to zero temperature, as is seen in Eq. (11). Supposing that a smaller value of k is taken into account, e.g., $k=0.1$, the second-order transition is expected to occur for $h<0.77$ (Fig. 3). In this situation the monotonic region near zero temperature becomes smaller and the topological quenching occurs more often, compared with the first-order case. The reason that the interval between quenching points in two situations decreases with increasing temperature is related to the behavior of the periodicity of $\overline{\tau_p}$ as a function of E in Eq. (11). In the thermal activation regime the phase

Φ vanishes because ϕ_3 in Eq. (8) approaches $\pi/2$, and thereby the oscillation or topological quenching does not exist any more.

In conclusion, we have considered oscillation of the tunnel splitting induced by temperature due to topological phase coherence in the biaxial spin system with a magnetic field along the hard axis. We have found that the topological quenching of the tunnel splitting occurs with increasing temperature, which is more prevalent in the biaxial nanomagnets with a lower value of k, i.e., with larger basal anisotropy. This feature is expected to be observable in nanomagnets including Fe_8 molecular magnets.

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[12] In the remaining field range the shape of the potential changes significantly and thereby the discussion becomes complicated at nonzero temperature, which will be presented elsewhere.

[13] Even though there will be dissipation due to the coupling of the system to the environment such as phonon at finite temperature, it will not change the main results discussed in this work. According to Ref. 4(e), the superohmic dissipation with the spectral density $J(\omega)\sim\omega^3$ induced by the phonon will be enhanced to $S_E^{\mathrm{eff,R}}$ in Eq. (9) with thermal dependence ($\sim T^4$) and dissipation coupling constant (Ref. 14) whose magnitude is expected to be small for $T<T_c$. Since the dissipation is weak, we can use the classical path found in its absence. This implies that, using the level splitting given by $\Delta E_n=[\omega(E_n)/\pi]\exp[-S_E(E_n)/2]$ in the WKB approximation, where ω is the attempt frequency in the well, Eqs. (8) and (9) will not change. However, it is of interest to ask how robust the topological oscillations are with respect to various dissipations even at $T=0$. Its quantitative study will be done elsewhere.

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